SF 298 MASTER COPY   KEEP THIS COPY FOR REPRODUCTION PURPOSES

REPORT DOCUMENTATION PAGE	Form Approved OMB NO. 0704-0188

1. AGENCY USE ONLY (Leave blank)	2. REPORT DATE	3. REPORT TYPE AND DATES COVERED
	August 30, 1999	Final Progress, 6/1/95 - 5/31/99

4. TITLE AND SUBTITLE
Coherent laser-atom interactions with partially coherent laser fields

5. FUNDING NUMBERS
DAAH04-95-1-0361

6. AUTHOR(S)
Daniel S. Elliott

7. PERFORMING ORGANIZATION NAMES(S) AND ADDRESS(ES)
Purdue University
1285 Electrical Engineering
West Lafayette, IN 47907

8. PERFORMING ORGANIZATION REPORT NUMBER

9. SPONSORING / MONITORING AGENCY NAME(S) AND ADDRESS(ES)
U.S. Army Research Office
P.O. Box 12211
Research Triangle Park, NC 27709-2211

10. SPONSORING / MONITORING AGENCY REPORT NUMBER
ARO 33488.4-PH

11. SUPPLEMENTARY NOTES
The views, opinions and/or findings contained in this report are those of the author(s) and should not be construed as an official Department of the Army position, policy or decision, unless so designated by other documentation.

12a. DISTRIBUTION / AVAILABILITY STATEMENT
Approved for public release; distribution unlimited.

12 b. DISTRIBUTION CODE

19991103 081

13. ABSTRACT *(Maximum 200 words)*

In this research program, the investigators have explored the role of fluctuations of a laser field on coherent interactions between the laser field and simple atomic systems. A key feature of these experiments is their ability to provide measurements of complex nonlinear optical interactions which are sufficient for direct, quantitative comparison with theory. The main thrust of the present work has been the study of non-Markovian field statistics, present in experiments that use multiple, time-delayed input beams to drive the nonlinear system. The specific interaction studied was Phase-Conjugate Four-Wave Mixing in an optically-pumped beam of atomic sodium, which the researchers measured as a function of laser bandwidth, Rabi frequency, and delay time between the pump and probe beams. The investigators have also calculated the expected signal strength through numerical integration of the Optical Bloch Equations. Experimental and numerical results are in very good agreement. The investigators also studied theoretically two-photon absorption with correlated, time-delayed input fields, and provided the first experimental confirmation of the fundamental theory of Phase-Conjugate Four-Wave Mixing with monochromatic fields in a simple atomic system.

14. SUBJECT TERMS
Coherent Laser-Atom Interactions, Four-wave Mixing, Laser Partial-coherence effects

15. NUMBER IF PAGES
6

16. PRICE CODE

17. SECURITY CLASSIFICATION OR REPORT	18. SECURITY CLASSIFICATION OF THIS PAGE	19. SECURITY CLASSIFICATION OF ABSTRACT	20. LIMITATION OF ABSTRACT
UNCLASSIFIED	UNCLASSIFIED	UNCLASSIFIED	UL

1 Statement of the Problems Studied:

During the period of this program, we have completed three projects, each resulting in a journal article, and made significant headway on a fourth project. The three completed projects include 1) the calculation of and critical comparison between the two-photon absorption spectra by a simple atomic system for different statistical models of cw laser fields, 2) the first experimental verification of the fundamental theory for phase-conjugate four-wave mixing with narrowband, cw laser fields in a nearly-Doppler free two-level atomic system, and 3) the first experimental observations of four-wave mixing with correlated, time-delayed, controlled stochastic fields. In the final project, currently underway, we are extending the measurements of the third project to include the more complex and theoretically challenging situation of correlated time-delayed intense fields. In this report we will describe each of these projects and the significance of each.

1.1 Two-photon absorption spectra for statistical laser fields

Over the past many years, two-photon absorption from fluctuating laser fields by simple atomic systems has been studied experimentally and theoretically. The spectra for these interactions, measured by recording the excitation probability as the central frequency of the optical field is tuned through the two-photon resonance frequency, showed several interesting behaviors. For example, the bandwidth of the absorption spectrum for absorption from the phase diffusing field (in the limit of the Lorentzian lineshape) increased at a rate of four times the bandwidth of the laser. A qualitative argument supporting this effect can be posed by considering the correlation time of the square of the optical field driving this transition. (The square of the field amplitude is the relevant quantity since this factor appears in the transition amplitude for a two-photon interaction.) Conversely, it can be shown experimentally and theoretically that the bandwidth of the two-photon absorption spectrum for absorption from the real Gaussian field (an optical field whose amplitude displays random, but real, Gaussian fluctuations) tends toward the natural linewidth of the transition, even in the limit of laser linewidths much larger than the absorption width. Again, this curious result can be explained in terms of the correlation properties of the two-photon moment. As part of the present program, the PI collaborated with Prof. Murray Hamilton at the University of Adelaide to calculate these lineshapes for a quartet of statistical field models of interest.

1.2 Phase-Conjugate Four-Wave Mixing in a nearly-Doppler-free Two-Level Atomic Medium

In our second set of investigations, we carried out a series of measurements of Phase-Conjugate Four-Wave Mixing (PCFWM) in a nearly-Doppler-free Two-Level Atomic Medium. Our primary purpose of these measurements was related to our interest in the effect of the partial coherence of the laser field on this important coherent atomic

interaction. We were surprised to discover, however, that there have been no experimental measurements which test the famous theoretical results of Abrams and Lind [R. L. Abrams and R. C. Lind, Optics Letters **2**, 94 (1978); **3**, 205 (1978)] for this interaction. In this theoretical work, the authors were able to calculate the PCFWM signal resulting from the interaction of a cw, monochromatic, stabilized laser field with a simple two-level absorber. PCFWM may result from the interaction of the medium with two counter-propagating beams, known as the forward and backward pump beams, and a weak probe beam which progagates in a direction at a small angle with respect to the forward pump beam. When all three input beams are degenerate, the phase-conjugate beam which results from this nonlinear interaction propagates precisely counter to the direction of the probe, and is also at the same frequency.

Many investigators have exploited this interaction for a variety of purposes, ranging from measurements of fast relaxation times in condensed-phase or vapor-phase media to generation of abberation-corrected reflections. Yet a test of the fundamental theory has been lacking since its first introduction in 1978. Without exception, all previous works concerning this interaction have been carried out in complex media (condensed phase, Doppler-broadened vapor-phase media, or multilevel systems) and/or with laser sources whose spectral width is broad. Our recent work, carried out with a cw frequency-stabilized tunable dye laser and an optically-pumped two-level atomic beam configuation, provides a direct quantitative confirmation of the Abrams and Lind theory.

1.3 Phase-Conjugate Four-Wave Mixing with Partially-Coherent Laser Fields

In our next investigations we explore the role of fluctuations of the laser phase and frequency in Phase-Conjugate Four-Wave Mixing (PCFWM). It is reasonable to expect that these field fluctuations should have a pronounced effect, as the interaction of the atom with the laser establishes coherences in the atomic system, and these atomic coherences drive the generation of the phase-conjugate field. Fluctuations of the laser field modify the coherence properties of the field, as well as the coherence properties that the laser field transfers to the atom. This is the basis for understanding the role of field fluctuations on PCFWM qualitatively. For the theorists, this interaction presents a difficult challenge due to the non-Markovian property of the total laser field. Briefly stated, a non-Markovian field is one in which there exist long term correlations. The net laser field in the present experiments is non-Markovian due to the temporal delay of the probe beam with respect to the pump.

There is considerable practical importance to these measurements as well. Four-wave mixing, and the closely-related nonlinear optical effect known as photon echo generation, have been important tools in measuring dephasing lifetimes in many media, vapor phase and condensed phase alike. It was discovered 15 years ago that broadband, nanosecond (or even cw) incoherent optical pulses could be used in these measurements, achieving temporal resolutions of picoseconds or shorter. The key to understanding this effect is to

realize that the limitation to measuring coherent interactions is not the pulse duration of the laser source, but rather the coherence time of the optical field. This effect has been exploited in a number of measurements, and is attractive in that the generation of broadband, incoherent pulses is in many case much simpler than that of ultrashort mode-locked (femtosecond) pulses. It is the goal of our project to provide quantitative data in a highly-controlled experimental geometry that can provide stringent tests of the theoretical techniques which have been developed to model this interaction.

1.4 Phase-conjugate reflectivity with partially-correlated pump beams

The problem of four-wave mixing with partially-correlated beams becomes much more complex when the two intense pump beams are only partially correlated. In the previously described work, we intentionally keep the delay between the forward and backward pump beams very small, so that the statistical fluctuations of these beams are the same. Thus the atomic coherences induced by the strong pump beams (forward and backward combined) are probed by the weak temporally-delayed probe beam. By delaying one of the pump beams, however, the net intense beam is non-Markovian, and the atomic response very complex. This additional degree of field decorrelation requires greater theoretical sophistication, and provides more stringent tests of techniques developed to calculate these fluctuating-field interactions. It is this situation which we are now studying with our experimental measurements and numerical techniques.

2 Summary of the Most Important Results:

2.1 Two-photon absorption spectra for statistical laser fields

The target of our attention in these investigations was the detailed spectral shape of the two-photon absorption spectrum for a series of different laser noise models. In particular, we systematically studied a periodic modulation, which became evident in the spectral wings, when a temporal delay was introduced between the two counter-propagating, fluctuating laser fields that drive the transition. The origin of these modulations is the partial decorrelation of the two laser fields due to their finite bandwidth and the induced delay. The models we studied include the Phase Diffusion Model, the Random Telegraph Model, the Complex Gaussian Model, and the Real Gaussian Model. While we observed the modulation for all laser statistical models studied, the modulation appears to be most obvious for the case of the Random Telegraph Phase noise when the magnitude of the phase jump is $\pi/4$. For this jump in the optical phase, the effect on the two-photon transition amplitude is maximized, and the sudden jump in phase, characteristic of this noise model, is large.

2.2 Phase-Conjugate Four-Wave Mixing in a nearly-Doppler-free Two-Level Atomic Medium

The results of our PCFWM measurements were in general in excellent agreement with those of the Abrams and Lind theory. In our experiments, the nonlinear medium is an optically-pumped beam of atomic sodium, and the laser source is a frequency-stabilized, cw, tunable, dye laser. We measured the peak reflectivity and the spectral linewidth of the interaction as a function of the intensity of the pump beams. The peak reflectivity, which we were able to calibrate on an absolute basis, agrees very nicely with theory for all intensities above the saturation intensity of the medium. For lower intensities, we discovered that the small degree of Doppler broadening resulting from the divergence of our atomic beam led to a significant decrease in the phase conjugate reflectivity. This effect is remarkable, in that the Doppler broadened absorption width of the transition (15 MHz) in our atomic beam is only slightly broader than the natural linewidth of the transition (10 MHz). Through numerical calculations, we were able to show good agreement between theory and experiment at these lower intensities as well. Bandwidths of the measured reflection spectra were also in excellent agreement with theory. It is noteworthy to remark that we were able to record the PCFWM signal using only 2 microwatts of probe power and a few milliwatts of pump power and atomic beam densities of only $3 \times 10^8 cm^{-3}$.

2.3 Phase-Conjugate Four-Wave Mixing with Partially-Coherent Laser Fields

These measurements are carried out with an apparatus very similar to that used for the measurements described above with the monochromatic field. The most significant difference is that we impose phase and frequency fluctuations onto the output of the stabilized laser source. The forward and backward pump beams and the probe beam are each derived from this source, ensuring that the phase/frequency fluctuations are identical on each. We then delay (or advance) the probe beam so as to invoke a partial decorrelation between the probe beam and the pump beam fluctuations. We vary the Rabi frequency of the interaction, the bandwidth of the laser spectrum and the delay time between the pump and probe, and measure the phase conjugate spectrum by tuning the laser frequency through the resonance frequency of the atomic transition. We supplement our experimental observations with results of numerical integration of the Optical Bloch Equations for this interaction. Some general observations regarding our results follow:

1) The phase-conjugate reflectivity is, in all cases, decreased from what it would be in the absence of the laser fluctuations.

2) The maximum PCFWM signal is generated when the delay between the pump and probe beams is nearly zero. As we implied above, this result can be argued on the basis of decay times. A large delay time between the pump and probe would allow the atomic coherences set up by the pump to die away before the correlated probe beam arrives.

Upon close examination, the numerical results actually reveal that a slight negative delay (typically about 3 nsec) yields the true maximum. This result is significantly harder to support intuitively, but is in agreement with similar theoretical and laboratory observations of related interactions.

3) Upon delaying the probe beam, the PCFWM signal decreases rapidly, with the most rapid decay corresponding to large Rabi frequencies. For large Rabi frequency, the signal tends to recover with further increase in the delay time. Both these effects seem to be related to the precession of the Bloch vector set up through the interaction.

4) With decreasing pump intensity (and Rabi frequency) the phase conjugate reflectivity decreases rapidly. We can understand this approximately in that at low intensities the system is driven only weakly, and the spectral overlap of the laser spectrum with that of the absorption decreases the strength of the interaction. At higher intensities, such that the system is strongly driven, the Rabi frequency becomes more important than the laser bandwidth and the decay rate of the medium, and the reflectivity reaches maximum values.

While the agreement between the experimental results and the numerical simulations have been very good, we are hopeful that these results will attract the attention of one of several theoretical groups who have the skills to attack these problems analytically. This problem poses a considerable challenge in that the interaction is non-Markovian due to the correlation between the pump and probe fields, and will involve development of new theoretical tools for its analysis.

2.4 Phase-conjugate reflectivity with partially-correlated pump beams

We are still recording and measuring spectra for this work, but preliminary results appear to be consistent with those of our numerical techniques. Further data collection and analysis is required before we are able to summarize these results.

3 List of publications and technical reports:

- M. W. Hamilton and D. S. Elliott, "Second order interference in two photon absorption," J. Mod. Optics **43**, 1765 (1996).

- Binh Do, Jongwhan Cha, D.S. Elliott and S.J. Smith, "Degenerate phase-conjugate four-wave mixing in a nearly-Doppler-free two-level atomic medium," Phys. Rev. A **58**, 3089 (1998).

- Binh Do, Jongwhan Cha, D.S. Elliott and S.J. Smith, "Phase-conjugate four-wave mixing with partially-coherent laser fields," Phys. Rev. A **60**, 508 (1999).

- Conference presentation: Binh Do and D.S. Elliott, "Experimental observations of degenerate four-wave mixing in Doppler-free two-level atoms by a phase diffusing

laser field," Annual Meeting of the Division of Atomic, Molecular, and Optical Physics, Santa Fe, NM, May 27-30, 1998.

4 Reportable Inventions:

None.

5 Participating Scientific Personnel:

- PI, Daniel S. Elliott
- Research Assistant, Binh Do
- Research Assistant, Jongwhan Cha, Ph.D. received May, 1997